



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<p>(21) International Application Number: PCT/IB99/01650  (22) International Filing Date: 8 October 1999 (08.10.99)  (30) Priority Data: 09/169,877 12 October 1998 (12.10.98) US  (71) Applicant (for all designated States except JP): CLARIANT FINANCE (BVI) LIMITED [---]; Citco Building, Wick- hams Cay, P.O. Box 662, Road Town, Tortola (VG).  (71) Applicant (for JP only): CLARIANT INTERNATIONAL LTD. [CH/CH]; Rothausstrasse 61, CH-4132 Muttenz (CH).  (72) Inventors: RUSSELL, Stephen, E.; 18730 Stonebridge Court, Grayslake, IL 60030 (US). WEBSTER, Joseph, R.; 10705 Summit Tree Court, Charlotte, NC 28277 (US).  (74) Agents: D'HAEMER, Jan et al.; Clariant International Ltd., Rothausstrasse 61, CH-4132 Muttenz (CH).</p>		<p>(81) Designated States: AU, CA, JP, KR, MX, ZA, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).  Published With international search report.</p>
<p>(54) Title: SOLID, SHAPED AND COLORED WAX ARTICLES</p> <div style="text-align: center; margin: 20px 0;"> <p>(1)</p> </div> <p>(57) Abstract</p> <p>Disclosed are improved solid, colored wax-based articles having longer color shelf-life. The invention comprises a natural and/or synthetic wax, a colorant, and an effective amount of a compound of formula (1) having symbols defined in the text. The preferred embodiments of the articles contain formula (1) that exhibits a UV absorptivity greater than or equal to 70 liters per gram-cm at a wavelength of less than or equal to 400 nm, as measured in either methanol, toluene or chloroform, using a conventional UV spectrophotometer. The uses for the articles include candles, crayons, fertilizer coatings, rosebush coatings, plant grafting, sealants, sun-cracking protection of rubber and plastic products, paper coating, packaging food products, electrical insulation, waterproofing, and precision investment casting.</p>		

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## SOLID, SHAPED AND COLORED WAX ARTICLES

### FIELD OF THE INVENTION

The field of the invention pertains to colored, wax-based articles such as crayons, paper coatings, food packaging, candles, and the like.

### BACKGROUND OF THE INVENTION

The term "wax", used herein is defined in accordance with the German Society of Fat Science as:

- (1) at 20°C it forms a solid whose consistency goes from kneadable to brittle;
- (2) having a macro- to -micro crystalline structure;
- (3) melting at temperatures of 40°C and above;
- (4) a relatively low viscosity at temperatures slightly above its melting point;
- (5) consistency and solubility that are strongly dependent upon temperature; and
- (6) that which can be polished under slight pressure.

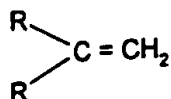
Among the various types of natural waxes are insect and animal waxes such as beeswax, lanolin, shellac wax, Chinese insect wax, and spermaceti; vegetable waxes such as carnauba, candelilla, Japan wax, ouricury wax, rice-bran wax, jojoba wax, castor wax, peat wax, petroleum waxes including petrolatum, paraffin wax, semi-microcrystalline wax, and microcrystalline wax, ozokerite and ceresin waxes; and the synthetic waxes such as polyethylene wax, Fischer-Tropsch wax, chlorinated naphthalene wax, chemically modified wax, substituted amide wax, ester waxes, hydrogenated vegetable fats and derivatives, alpha olefins and polymerized alpha olefin wax.

Waxes are used in a variety of applications including, but not limited to, polishes, candles, crayons, fertilizer coatings, rosebush coatings, plant grafting, sealants, sun-cracking protection of rubber and plastic products, paper coating, packaging food products electrical insulation, waterproofing and cleaning compounds, carbon paper, and precision investment casting.

Some polymers of higher alpha olefins, for example  $C_{20}$ , are waxes and are sold as synthetic waxes. The polymerization process yields highly branched materials, typically with

broad molecular weight distributions. Properties of the individual products are highly dependent on the alpha olefin monomers and polymerization conditions. The molecular structure of poly( $\alpha$ -olefins) is sometimes used to modify the properties of paraffin wax, primarily for improving shape retention in unsupported, molded or dipped articles, such as candles. The products can increase the hardness and opacity of the paraffin, without increasing the cloud point or viscosity.

U.S. Patent No. 4,060,569 relates to hydrocarbon polymers having lower penetration or greater hardness, greater viscosities, higher molecular weights, but having melting points and congealing points which are essentially no higher, but preferably lower than the original hydrocarbon. U.S. Patent No. 4,239,546 relates to the use of free-radical polymerized  $\alpha$ -olefin hydrocarbons to improve the hardness of waxes. In both patents, the hydrocarbons employed are primarily alpha olefins of the formula  $RCH=CH_2$ , but also include alpha olefins of the vinylidene structure



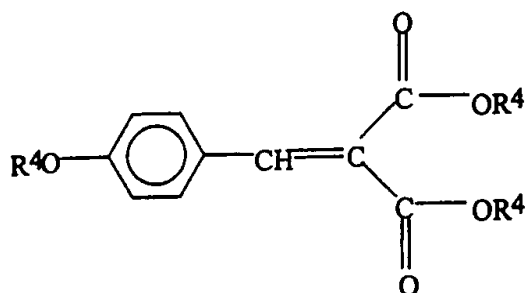
or a mixture of alpha olefins, vinylidenes, internal olefins and saturates.

UV light is known to break chemical bonds in colorants and waxes, resulting in loss of original properties. An organic mixture or compound, such as a colorant or wax, can also degrade through free radical attack. UV absorber can be used to dissipate the absorbed energy as heat. The UV stabilization of pigmented waxes containing free-radical polymerized alpha olefin wax absorbers presents a problem from the standpoint of interference between the  $\alpha$ -olefin and UV stabilizer. The loss of intensity of shade of colorants such as dyes and pigments is pronounced due to strong UV light absorption by the colorant.

There still exists a need to provide the hardness for wax-based articles for dimensional stability, and yet still a need for improved resistance to color-fading while exposed to fluorescent light or ultraviolet (UV) light. Conventional UV absorbers used for UV stabilization of waxes are the hydroxy substituted benzophenones and benzotriazoles. These compounds differ in cost, solubility, volatility, and their absorptivity at specific wavelengths, but show no remarkable properties in colored wax-based articles.

The use of UV absorbers in cosmetic compositions for human skin and hair applications is known. U.S. Patent No. 5,508,025 discloses UV absorbers and cosmetic compositions containing a naphthalene methylene malonic diester.

The use of UV absorbers in plastics and coatings to enhance weather resistance is well known. U.S. Patent 4,301,209 discloses a radiation curable coating composition comprising a benzylidene malonic acid ester UV light absorber of the formula



wherein  $R^4$  is independently alkyl or hydroxyalkylene.

It is known from U.S. Patent 4,260,732 and 4,404,257 to stabilize polycarbonate resins for improved resistance to UV radiation with UV absorber (UVA) compounds including the benzophenone derivatives, benzotriazole derivatives, benzoate esters, phenyl salicylates, derivatives of crotonic acid, malonic acid esters, and cyanoacrylates. U.S. Patent No. 5,439,958 discloses piperidine compounds which can be used as light stabilizers, heat stabilizers, and oxidation stabilizers for organic materials, in particular synthetic polymers.

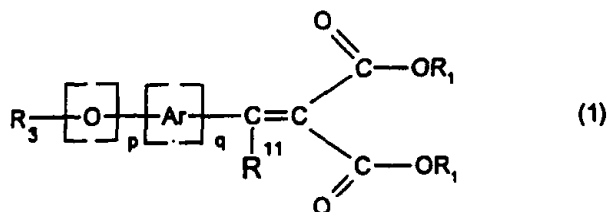
The thermal degradation is a concern primarily during manufacturing processes where a wax is held for extended lengths of time at or above the melt temperature. Antioxidants are often added to the melted wax to inhibit degradation during manufacturing. The melting point of thermally degraded candle wax is lower than the original wax material. The thermally degraded material also becomes more UV light sensitive and accelerates the photodegradation of dyes.

The problem of rapid color fading is manifested in finished candles which are displayed at the point of purchase under exposure to sunlight and/or fluorescent light. Even fluorescent light (wavelength from 290 to 380 nm) causes conventional colored candles to fade noticeably in a short period of time. Premature color fading is therefore problematic for decorative wax

articles and there is a continuing need for longer shelf-life yet there is no basis for predicting improved stabilizing effects in relatively low molecular weight, nonpolar waxes based on observations of effects seen among the stabilizers used for high polymers which are polar in nature. A surprising improvement in the color retention and shelf life of wax-based articles has been observed with a particular UV absorber. The UV absorbing additives which show the improvement exhibit a UV absorptivity greater than or equal to 40 liters, especially greater than 70 liters, per gram-cm. at a wavelength of less than or equal to 400 nm, as measured in either methanol, toluene or chloroform, using a UV spectrophotometer.

### SUMMARY OF THE INVENTION

It is an object of the present invention to produce an improved, solid, colored, molded wax-based article having surprisingly longer color shelf-life. The invention comprises a synthetic wax, a colorant, and an effective amount of a compound of the formula (1)



wherein

$\text{R}_1$  is independently  $\text{C}_{1-20}$  alkyl,  $\text{C}_{1-20}$  alkoxyalkyl,  $\text{C}_{1-20}$  hydroxyalkyl,  $\text{C}_{1-20}$  alkenyl, substituted  $\text{C}_{1-20}$  alkenyl groups,  $\text{C}_{1-20}$ -alkoxy- $\text{C}_{1-20}$ -alkyl groups,  $\text{C}_{1-20}$ -oxy-N- $\text{C}_{1-20}$ -alkyl groups, -N-cyclic- $\text{C}_{1-10}$ -alkyl groups, and cyclic-N- $\text{C}_{1-10}$ -alkyl groups;

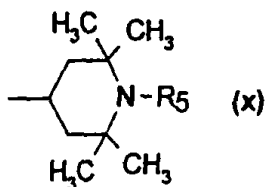
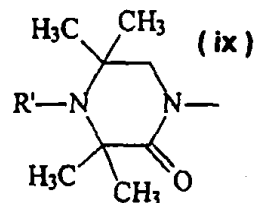
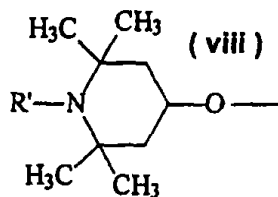
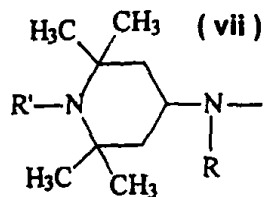
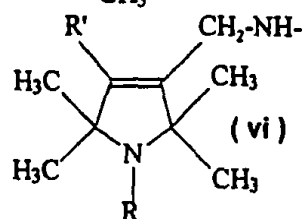
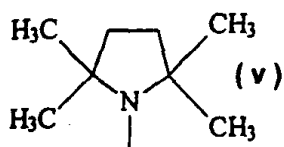
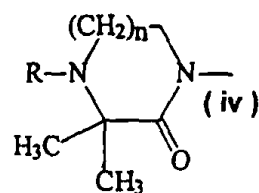
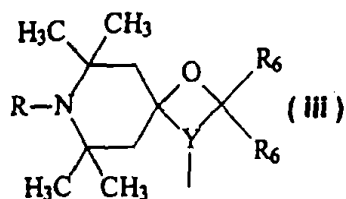
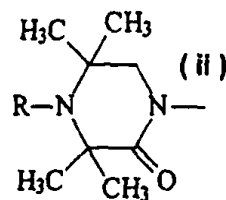
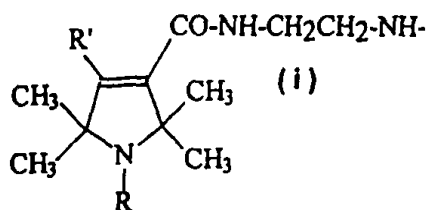
$\text{R}_{11}$  is hydrogen or  $\text{C}_1\text{-C}_8$  alkyl;

p and q are independently 0 or 1;

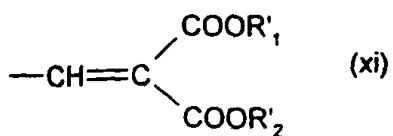
Ar is a substituted or unsubstituted aromatic single ring or a substituted or unsubstituted aromatic fused 2 or 3 ring group or a heteroaromatic single ring or a heteroaromatic fused 2 or 3 ring group, for example a substituted or unsubstituted phenyl or naphthyl group;

$\text{R}_3$  is hydrogen,  $\text{C}_1\text{-C}_{18}$  alkyl,  $\text{C}_5\text{-C}_{12}$  cycloalkyl unsubstituted or mono-, di- or tri-substituted by  $\text{C}_1\text{-C}_4$  alkyl, or  $\text{C}_7\text{-C}_9$  phenylalkyl unsubstituted or mono-, di- or tri-

substituted on the phenyl by C<sub>1</sub>-C<sub>4</sub>alkyl, or preferably a hindered amino group  
 selected from (i) to (x).



or a group (xi):



wherein

R, R', R<sub>1</sub>', R<sub>2</sub>' and R<sub>5</sub> are independently either hydrogen, C<sub>1-12</sub>alkyl, C<sub>1-8</sub>alkoxy, or -COR<sub>3</sub>, where R<sub>3</sub> is hydrogen, C<sub>1-8</sub>alkyl, phenyl, -COO(C<sub>1-4</sub>alkyl) or NR<sub>15</sub>R<sub>16</sub>, where R<sub>15</sub> and R<sub>16</sub> are independently hydrogen, C<sub>1-12</sub>alkyl, C<sub>5-8</sub>cycloalkyl, phenyl or (C<sub>1-12</sub>alkyl)phenyl; or R<sub>15</sub> and R<sub>16</sub> together with the N-atom to which they are attached form a five- to seven-membered ring which may contain an additional N- or O-atom (preferably forming a piperidine or morpholine ring);

n is 0 or 1 (structure iv);

Y is the group -NCO or -OCN, where CO forms part of the cyclic structure;

each R<sub>6</sub> of structure (iii) independently is selected from hydrogen, C<sub>1-12</sub>alkyl or phenyl provided only one R<sub>6</sub> can be phenyl, or both groups R<sub>6</sub> together form the group -(CH<sub>2</sub>)<sub>m</sub>-; where m is 2 to 11, -C(CH<sub>3</sub>)<sub>2</sub>-, -C(CH<sub>3</sub>)<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>- or -C(CH<sub>3</sub>)CH<sub>2</sub>CH<sub>2</sub>CH(CH<sub>3</sub>)-. Preferably R is hydrogen, C<sub>1</sub>-C<sub>8</sub>alkyl, O, OH, CH<sub>2</sub>CN, C<sub>1</sub>-C<sub>18</sub>alkoxy, C<sub>5</sub>-C<sub>12</sub>cycloalkoxy, or C<sub>3</sub>-C<sub>6</sub>alkenyl, C<sub>7</sub>-C<sub>9</sub>phenylalkyl unsubstituted or mono-, di- or tri-substituted on the phenyl by C<sub>1</sub>-C<sub>4</sub>alkyl; or aliphatic or aromatic C<sub>1</sub>-C<sub>10</sub>acyl;

R'<sub>1</sub> and R'<sub>2</sub> in (xi) are selected independently from C<sub>1-20</sub> alkyl, C<sub>1-20</sub>alkoxyalkyl, C<sub>1-20</sub> hydroxyalkyl, C<sub>1-20</sub> alkenyl, substituted C<sub>1-20</sub> alkenyl groups, C<sub>1-20</sub>-alkoxy-C<sub>1-20</sub>-alkyl groups, C<sub>1-20</sub>-oxy-N-C<sub>1-20</sub>-alkyl groups, -N-cyclic-C<sub>1-10</sub>-alkyl groups, and cyclic-N-C<sub>1-10</sub>-alkyl groups, such as (I) to (x) above.

The solid, colored, wax-based article of the present invention surprisingly has improved resistance to color loss from exposure to UV light and sunlight in combination with an  $\alpha$ -olefin wax which has residual peroxide of less than 200 ppm, preferably less than 100 ppm, and most preferably less than 50 ppm. The time until a noticeable loss of color intensity is many-fold longer compared to the state of the art for colored wax-based articles.

It is also an object of the present invention to produce a solid, colored, molded candle in a variety of shapes, sizes, and forms that has improved shelf-life that is more desirable to the consumer.



It is a further object of the invention to provide a wax concentrate consisting essentially of a natural and/or synthetic wax and from about 0.02% to 5% by weight, preferably 0.05% to 1%, and more preferably from 0.1% to 0.3% by weight of a compound according to (1) based on the total weight of the wax:

#### DESCRIPTION OF THE PREFERRED EMBODIMENTS

The wax-based articles comprise at least 75% by weight of a natural and/or synthetic wax, a coloring amount of colorant, and a compound of the formula (1) is essential in the present invention and takes the form of a solid, colored, shaped wax-based article that has distinct advantages over existing solid, colored, shaped wax-based articles. In particular, the article of the present invention has unexpected long color life underexposure to UV light. The article can be prepared in any desired shape, size, or form. One such embodiment is a candle comprising a wick extending within the mass of wax.

Dyes and pigments are among the colorants that may be used in the solid, colored, shaped wax-based article of the present invention. Fluorescent colorants can also be used as colorants in the present invention. Particular preference is given to oil soluble dyes. Oil soluble dyes that can be used as colorants in the present invention include, but are not limited to

C.I. Solvent Yellow 3, C.I. Solvent Green 3, C.I. Acid Black 429, C.I. Solvent Red 207, C.I. Phthalo Blue, C.I. Solvent Violet 13, C.I. Disperse Violet 17, C.I. Solvent Yellow 14, C.I. Solvent Blue 36, C.I. Solvent Yellow 33, C.I. Solvent Red 149, C.I. Solvent Yellow 56, C.I. Solvent Yellow 90, C.I. Solvent Red 26, C.I. Solvent Yellow 29, C.I. Solvent Red 24, C.I. Solvent Yellow 16, C.I. Solvent Yellow 72, C.I. Solvent Blue 104, C.I. Solvent Yellow 93, C.I. Solvent Yellow 98, C.I. Disperse Yellow 64, C.I. Solvent Yellow 24, C.I. Solvent Orange 60, C.I. Vat Red 41, C.I. Solvent Red 195, C.I. Solvent Red 111, C.I. Solvent Red 179, C.I. Solvent Red 135, C.I. Solvent Red 63, C.I. Disperse Violet 26, C.I. Solvent Violet 37, C.I. Solvent Green 28, C.I. Solvent Yellow 133, C.I. Solvent Yellow 164, C.I. Solvent Blue 122, C.I. Solvent Violet 49, C.I. Solvent Brown 122, C.I. Solvent Red 91, C.I. Solvent Red 164, C.I. Solvent Blue 98

Pigments can also be used as colorants in the present invention. Particularly preferred are pigments that can act as oil soluble dyes which include, but are not limited to C.I. Pigment Yellow 74, C.I. Pigment Yellow 192 and families thereof which are available in the art.

A blend of paraffin wax and synthetic wax can be used in the present invention. The natural waxes include, but are not limited to the aforementioned insect and animal waxes such as beeswax, lanolin, shellac wax, Chinese insect wax, and spermaceti; vegetable waxes such as carnauba, candelilla, Japan wax, ouricury wax, rice-bran wax, jojoba wax, castor wax, bayberry wax, sugar cane wax, and maize wax; mineral waxes such as montan wax, peat wax, petroleum waxes including petrolatum, paraffin wax, semimicrocrystalline wax, and microcrystalline wax; ozokerite, hydrogenated fats and derivatives, and ceresin waxes. The hydrogenated fats can be derived from soybean oil, palm oil, tallow and cottonseed oil, and the like which are commercially available.

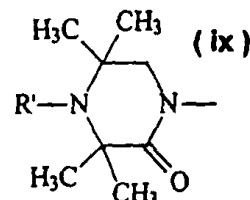
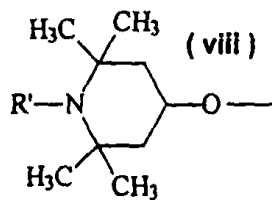
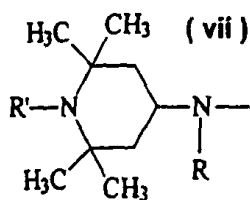
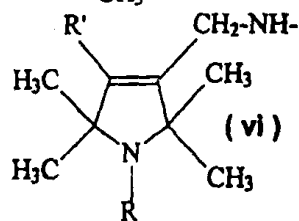
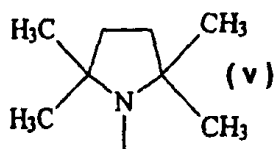
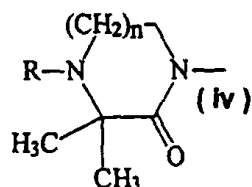
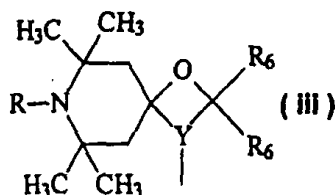
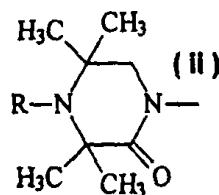
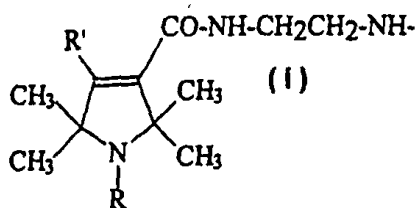
The synthetic waxes that may be used in the present invention include, but are not limited to, polyethylene wax (m.w. up to about 6000), Fischer-Tropsch wax, chlorinated naphthalene wax, chemically modified wax, substituted amide wax, ester waxes, polymerized alpha olefin wax; or any blend of the above. Some of these synthetic waxes are commercially available as Shell<sup>®</sup>100, Mobil<sup>®</sup>130, Astor<sup>®</sup>5212, monomeric  $\alpha$ -olefin, e.g. Chevron<sup>®</sup>C30, and polymerized  $\alpha$ -olefin, for example Vybar<sup>®</sup>103. A synthetic wax that is preferred in the present invention is a polymerized alpha olefin wax that has a active peroxide content of less than 200 ppm, preferably less than 100 ppm, and most preferably less than 50 ppm. More preferred are the synthetic waxes disclosed in U.S. Patent No. 4,060,569 and U.S. Patent No. 4,239,546, both of which are specifically incorporated herein by reference with the proviso that the residual peroxide content is in the above specified range. The most preferred is a alpha olefin wax wherein the alpha olefin is an  $\alpha$ -C<sub>24</sub>-C<sub>54</sub> hydrocarbon wax, which branched and/or linear, for example CAS No. 131459-42-2 which is commercially available as Chevron<sup>®</sup>C30.

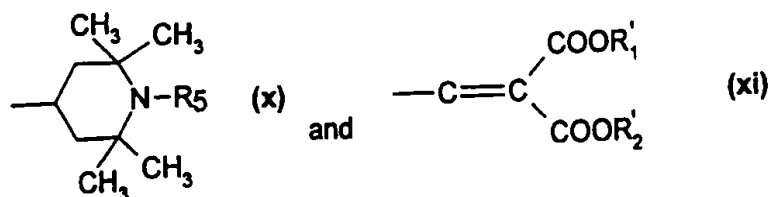
The solid, colored, molded wax-based article of the present invention comprises a natural and/or a synthetic wax, an effective amount of colorant, and the compound of the formula (1) which exhibits a UV absorptivity greater than or equal to 40 liters per gram-cm. at a wavelength of less than or equal to 400 nm, as measured in either methanol, toluene or chloroform, using a UV spectrophotometer; the absorptivity varies according to the selected substituents, wherein R<sub>1</sub> is preferably, independently C<sub>1-8</sub> alkyl, C<sub>1-6</sub> alkoxyalkyl,

C<sub>1-12</sub> hydroxyalkyl, C<sub>1-12</sub> alkenyl, substituted C<sub>1-16</sub> alkyl groups, C<sub>1-6</sub>-alkoxy-C<sub>1-12</sub>-alkyl groups; and R<sub>11</sub> is hydrogen or C<sub>1-4</sub> alkyl.

"Ar" is a substituted or unsubstituted aromatic single ring or a substituted or unsubstituted aromatic fused 2 or 3 ring group or a heteroaromatic single ring or a heteroaromatic fused 2 or 3 ring group, for example a substituted or unsubstituted phenyl or naphthyl group.

R<sub>3</sub> is hydrogen, C<sub>1</sub>-C<sub>18</sub>alkyl, C<sub>5</sub>-C<sub>12</sub>cycloalkyl unsubstituted or mono-, di- or tri-substituted by C<sub>1</sub>-C<sub>4</sub>alkyl, or C<sub>7</sub>-C<sub>9</sub> phenylalkyl unsubstituted or mono-, di- or tri-substituted on the phenyl by C<sub>1</sub>-C<sub>4</sub>alkyl, or preferably a hindered amino group selected from (I) to (x):





wherein

R, R', R'<sub>1</sub>, R'<sub>2</sub>

R<sub>5</sub>

R, R', R'<sub>1</sub>, R'<sub>2</sub> and R<sub>5</sub>

n

Y

each R<sub>6</sub>

are independently either hydrogen, C<sub>1-12</sub>alkyl, C<sub>1-8</sub>alkoxy;

can be either hydrogen, C<sub>1-12</sub>alkyl, C<sub>1-8</sub>alkoxy, or C<sub>1-10</sub> acyl;

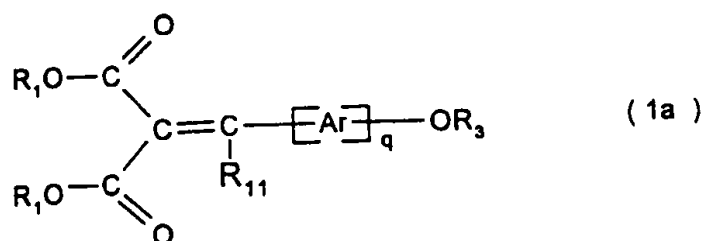
can be -COR<sub>3</sub>, where R<sub>3</sub> is hydrogen, C<sub>1-8</sub>alkyl, phenyl, -COO(C<sub>1-4</sub>alkyl) or NR<sub>15</sub>R<sub>16</sub>, where R<sub>15</sub> and R<sub>16</sub> are independently hydrogen, C<sub>1-12</sub>alkyl, C<sub>5-6</sub>cycloalkyl, phenyl or (C<sub>1-12</sub>alkyl)phenyl; or R<sub>15</sub> and R<sub>16</sub> together with the N-atom to which they are attached form a five- to seven-membered ring which may contain an additional N- or O-atom (preferably forming a piperidine or morpholine ring);

is 0 or 1 (structure iv);

is the group -NCO or -OCN, where CO forms part of the cyclic structure;

of structure (iii) independently is selected from hydrogen, C<sub>1-12</sub>alkyl or phenyl provided only one R<sub>6</sub> can be phenyl, or both groups R<sub>6</sub> together form the group -(CH<sub>2</sub>)<sub>m</sub>-, where m is 2 to 11, -C(CH<sub>3</sub>)<sub>2</sub>-, -C(CH<sub>3</sub>)<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>- or -C(CH<sub>3</sub>)CH<sub>2</sub>CH<sub>2</sub>CH(CH<sub>3</sub>)-. Preferably R is hydrogen, C<sub>1</sub>-C<sub>8</sub>alkyl, O, OH, CH<sub>2</sub>CN, C<sub>1</sub>-C<sub>18</sub>alkoxy, C<sub>5</sub>-C<sub>12</sub>cycloalkoxy, or C<sub>3</sub>-C<sub>6</sub>alkenyl, C<sub>7</sub>-C<sub>9</sub>phenylalkyl unsubstituted or mono-, di- or tri-substituted on the phenyl by C<sub>1</sub>-C<sub>4</sub>alkyl; or aliphatic or aromatic C<sub>1</sub>-C<sub>10</sub>acyl.

An alternative suitable compound according to formula (1) is (1a):



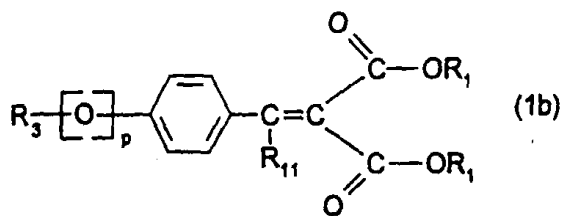
wherein

p is 1;

each  $R_1$  is independently  $C_{1-20}$  alkyl,  $C_{1-20}$  alkoxyalkyl,  $C_{1-20}$ hydroxyalkyl,  $C_{1-20}$  alkenyl, substituted  $C_{1-20}$  alkenyl groups,  $C_{1-20}$ -alkoxy- $C_{1-20}$ -alkyl, or selected from groups (i) to (x); and

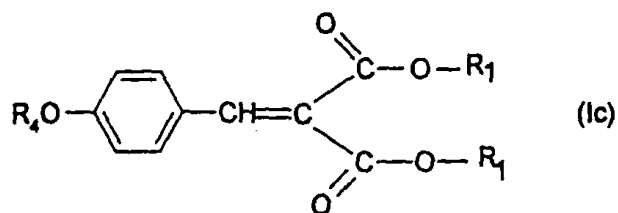
$R_3$ , q,  $R_{11}$ , and Ar are defined as above for (1).

With respect to formula (1), when  $q = 1$ , Ar is preferably phenyl and formula (1) has the following structure (1b):



wherein  $R_1$ ,  $R_3$ ,  $R_{11}$ , and p are defined as above for compound (1).

More preferably, p and q are each 1, Ar is phenyl, and the compounds of formula (1) are (1c):



wherein

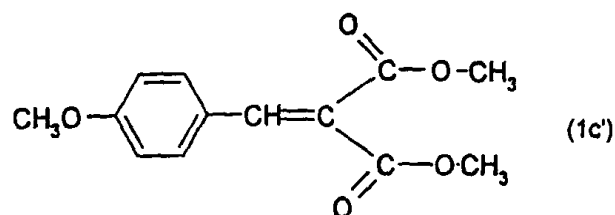
$R_1$  is linear or branched  $C_{1-10}$  alkyl;

$R_4$  is linear or branched  $C_{1-10}$  alkyl or  $C_{1-10}$  hydroxyalkyl. Particularly preferred for  $R_1$  and  $R_4$  are identical  $C_{1-6}$  alkyl groups, e.g., methyl, ethyl, propyl, butyl, pentyl and hexyl groups.

The preferred embodiments of (1) exhibit a UV absorptivity greater than or equal to 40 liters per gram-cm. at a wavelength of less than or equal to 400 nm, as measured in either methanol, toluene or chloroform, using a conventional UV spectrophotometer. More

preferred embodiments of (1) exhibit a UV absorptivity greater than or equal to 70 liters per gram-cm. at a wavelength of less than or equal to 400 nm, as measured in either methanol, toluene or chloroform, using a UV spectrophotometer. The most preferred embodiments of (1) exhibit a UV absorptivity greater than or equal to 90 liters per gram-cm. at a wavelength of less than or equal to 400 nm, as measured in either methanol, toluene or chloroform, using a UV spectrophotometer.

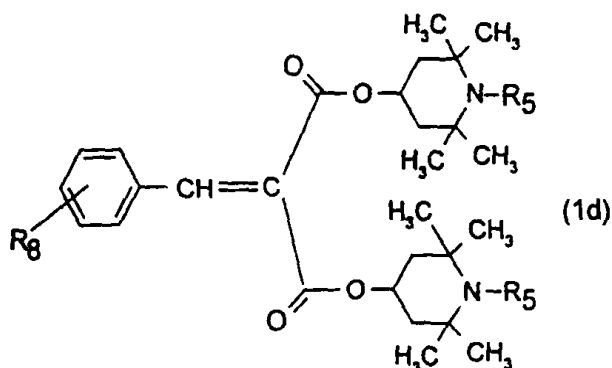
When  $p=1$ ,  $R_1=R_4=CH_3$ , formula (1) is the following most preferred formula (1c')



(absorptivity of 1c' is 105 l/gm-cm at  $\lambda < 400$  nm)

Specific examples of hydroxybenzylated malonates, include, for example dioctadecyl-2,2-bis(3,5-di-tert-butyl-2-hydroxybenzyl)malonate, di-octadecyl-2-(3-tert-butyl-4-hydroxy-5-methylbenzyl)malonate, di-dodecylmercaptoethyl-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)malonate, di[4-(1,1,3,3-tetramethylbutyl)-phenyl]-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)malonate.

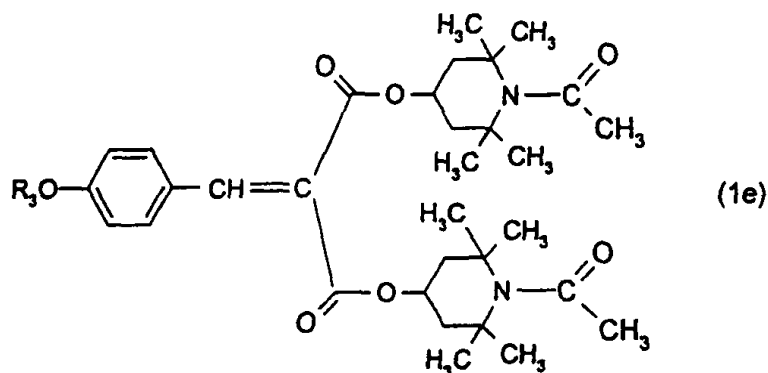
Other examples of formula (1) include compounds (1d) and (1e) containing one or more hindered amino groups (HALS). For example, the compound of the formula (1d) where  $q=1$  and  $Ar=phenyl$ :



wherein

$R_3$  is as defined as in (1) above;

$R_8$  is hydrogen,  $C_1 - C_4$  alkyl,  $C_1 - C_8$  alkoxy, and preferably  $R_8$  is para  $C_1 - C_8$  alkoxy, most preferably hydrogen, p-methoxy or p-ethoxy; and



wherein  $R_3$  is defined as in (1) above.

For  $R_3 = -CH_3$ , (1e) absorptivity is 55 l/gm-cm at  $\lambda < 400$  n.

Further examples of a composition comprising a natural and/or synthetic wax, a colorant, and stabilizer (1) are those in which the stabilizer selected from the following compounds:

bis(2,2,6,6-tetramethyl-4-piperidyl) [[3,5-bis(1,1-dimethylethyl)-4-hydroxyphenyl]methyl] malonate;

bis(1,2,2,6,6-pentamethyl-4-piperidyl)-2-(4-methoxybenzylidene)malonate;

bis[2,2,6,6-tetramethyl-1-(1-oxo-2-propenyl)-4-piperidinyl] ester;

propanedioic acid, [[3,5-bis(1,1-dimethylethyl)-4-hydroxyphenyl]methyl]butyl-bis(1,2,2,6,6-pentamethyl-4-piperidinyl)ester;

malonic acid, (3-tert-butyl-4-hydroxy-5-methylbenzyl)- dioctadecyl ester;

(p-methoxybenzylidene)malonic acid diethyl ester;

dimethyl (4-hydroxybenzylidene) malonate;

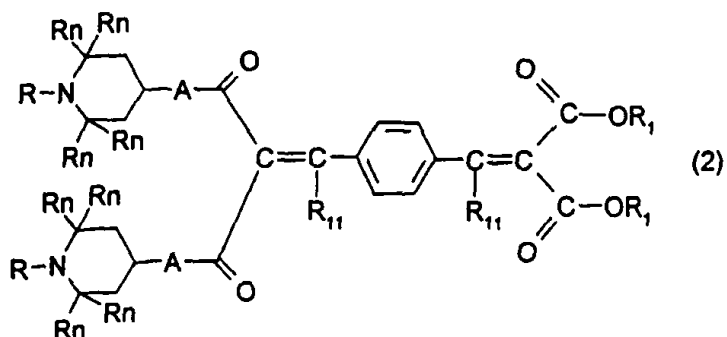
dimethyl (4-hydroxyphenyl) malonate;

di-isobutyl (p-methoxybenzylidene)malonate;

di-( $C_{1-20}$ alkyl)(4-phenylbenzylidene) malonate, e.g. dimethyl, diethyl, or dibutyl-(4-phenylbenzylidene) malonate; and

tetraethyl-2,2'-[methylenebis(4,1-phenyleneiminocarbonyl)]bismalonate.

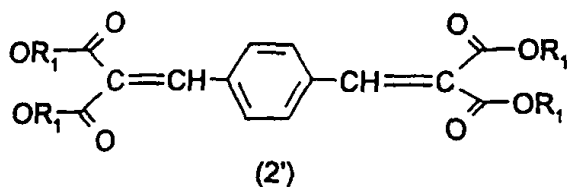
Suitable compounds of formula (1) when  $p = 0$  and  $q = 1$  are (2):



wherein

$R$ ,  $R_1$  and  $R_{11}$  are independently defined as for (1) above, and  
 each  $R_n$  is independently  $-CH_3$  or  $-CH_2(C_{1-4}alkyl)$  or adjacent  $R_n$  groups form a  
 group  $-(CH_2)_5-$ , and  
 $A$  is  $-O-$  or  $-N(C_{1-4}alkyl)-$  or  $-NH-$ , and preferably  $-O-$ .

Alternatively, suitable compounds of formula (1) when  $p = 0$  and  $q = 1$  can be (2')



wherein each  $R$ , is independently defined as for compound (1) above.

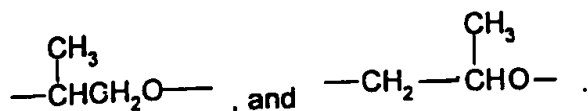
Examples of alkyl groups having not more than 20 carbon atoms are methyl, ethyl, propyl, isopropyl, butyl, 2-butyl, isobutyl, t-butyl, pentyl, 2-pentyl, hexyl, heptyl, octyl, 2-ethylhexyl, t-octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, hexadecyl and octadecyl; exemplary branched alkyl groups are isopropyl, isobutyl, t-butyl, 2,2-dimethylpropyl, 2-methylpropyl, cyclohexylmethyl, cyclohexylethyl, 2-ethylhexyl, 3,5,5-trimethylhexyl, 2-hexylundecyl, and 5,7,7-trimethyl-2-(1,3,3-trimethylbutyl)hexyl.



Exemplary cyclic alkyl groups are cyclohexyl-, and methylcyclohexyl-.

Examples of alkoxy groups are methoxy, ethoxy, propoxy, isopropoxy, butoxy, isobutoxy, pentoxy, isopentoxy, hexoxy, 2-ethylhexyloxy, heptoxy, octoxy, decyloxy, dodecyloxy, tetradecyloxy, hexadecyloxy and octadecyloxy, nonadecyloxy. Preferred alkoxy examples are C<sub>6</sub>-C<sub>12</sub>alkoxy, in particular heptoxy and octoxy.

Examples of the alkoxyalkylenoxyalkyl groups which are preferred are C<sub>1</sub>-20-alkoxy-C<sub>1</sub>-5-alkylenoxy-C<sub>1</sub>-20-alkyl groups. The C<sub>1</sub>-20-alkoxy groups and C<sub>1</sub>-20-alkyl groups include those mentioned as examples of the alkoxyalkyl groups. Examples of the C<sub>1</sub>-5-alkylenoxy groups include -CH<sub>2</sub>O-, -CH<sub>2</sub>CH<sub>2</sub>O-, -CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>O-.



Particularly, C<sub>1</sub>-12-alkoxy-C<sub>1</sub>-5-alkylenoxy-C<sub>1</sub>-12-alkyl groups are more preferred.

Examples of C<sub>5</sub>-C<sub>12</sub>cycloalkyl R<sub>3</sub> unsubstituted or mono-, di- or tri-substituted by C<sub>1</sub>-C<sub>4</sub>alkyl are cyclopentyl, methylcyclopentyl, dimethylcyclopentyl, cyclohexyl, methylcyclohexyl, dimethylcyclohexyl, trimethylcyclohexyl, t-butylcyclohexyl, cyclooctyl, cyclodecyl and cyclododecyl. Unsubstituted or substituted cyclohexyl groups are preferred.

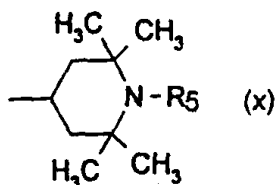
Examples of R<sub>3</sub> as C<sub>5</sub>-C<sub>12</sub> cycloalkoxy R<sub>1</sub> are cyclopentoxy, cyclohexoxy, cycloheptoxy, cyclooctoxy, cyclodecyloxy and cyclododecyloxy. Cyclopentoxy and cyclohexoxy are preferred.

Examples of C<sub>3</sub>-C<sub>6</sub> alkenyl are allyl, 2-methylallyl, butenyl and hexenyl, 3-butenyl, and 10-undecenyl; examples of branched alkenyl groups are 1-methyl-2-propenyl, 3-methyl-3-butenyl. Allyl is preferred.

Examples of C<sub>7</sub>-C<sub>9</sub> phenylalkyl unsubstituted or mono-, di- or tri-substituted on the phenyl by C<sub>1</sub>-C<sub>4</sub>alkyl are benzyl, methylbenzyl, dimethylbenzyl, trimethylbenzyl, t-butylbenzyl and 2-phenyl thyl. Benzyl is preferred.

Examples of aliphatic and aromatic C<sub>1</sub>-C<sub>10</sub> acyl are C<sub>1</sub>-C<sub>8</sub>alkanoyl or C<sub>3</sub>-C<sub>8</sub>- alkenoyl, for example, acetyl, propionyl, butyryl, isobutyryl, pentanoyl, benzoyl, hexanoyl, heptanoyl, octanoyl, 2-ethylhexanoyl, acryloyl and crotonyl. Acetyl is preferred.

Those compounds of the formula (1) are preferred in which R<sub>1</sub> is hydrogen or C<sub>1</sub>-C<sub>6</sub>alkyl and R<sub>3</sub> is C<sub>1</sub>-C<sub>16</sub>alkyl, C<sub>5</sub>-C<sub>8</sub>cycloalkyl unsubstituted or mono-, di- or tri-substituted by C<sub>1</sub>-C<sub>4</sub>alkyl; benzyl unsubstituted or mono-, di- or tri-substituted on the phenyl by C<sub>1</sub>-C<sub>4</sub>alkyl; or a group of the formulas (i) - (x) above, preferably (x).

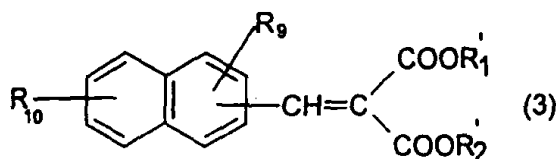


Preferable alkoxyalkyl groups represented by R<sub>5</sub> are C<sub>1</sub>-20-alkoxy-C<sub>1</sub>-20-alkyl groups, C<sub>1</sub>-20-oxy-N-C<sub>1</sub>-20-alkyl groups, especially C<sub>1</sub>-20-oxy-N-cyclic-C<sub>1</sub>-10-alkyl groups. Of C<sub>1</sub>-20-alkoxy groups, with C<sub>1</sub>-12-alkoxy groups more preferred among these.

Compounds of the formula (1) where q = 1 are prepared according to processes disclosed in U.S. Patent No. 5,705,545, incorporated herein by reference.

Compounds of the formula (1), where q = 0 are prepared according to processes disclosed in U.S. Patent No. 5,439,958, incorporated herein by reference. The most preferred additive in the shaped, colored wax article is available under the Sanduvor® mark, designated PR-25, from Clariant.

A specific example of formula (1) where Ar is naphthyl, is a compound represented by formula (3):

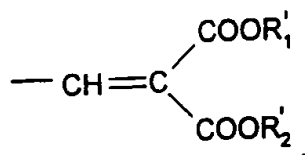


wherein

$R'_1$  and  $R'_2$  each independently represent C<sub>1-20</sub> alkyl, C<sub>1-20</sub> alkoxyalkyl, C<sub>1</sub>-hydroxyalkyl, C<sub>1-20</sub> alkenyl, substituted C<sub>1-20</sub> alkenyl groups, C<sub>1-20</sub>-alkoxy-C<sub>1-20</sub>-alkyl groups, C<sub>1-20</sub>-oxy-N-C<sub>1-20</sub>-alkyl groups, -N-cyclic-C<sub>1-10</sub>-alkyl groups, and cyclic-N-C<sub>1-10</sub>-alkyl groups, such as (I) to (x) above;

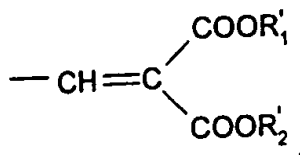
$R_{10}$  represents a hydrogen atom, an alkoxy group, an alkenyloxy group, a hydroxy group, an acyl group, an alkyl group, or an alkenyl group;

$R_9$  represents a hydrogen atom, an alkoxy group, an alkenyloxy group, a hydroxy group, an acyl group, an alkyl group, an alkenyl group, or



where  $R'_1$  and  $R'_2$  have the same meaning as above for (xi).

It is preferred that the alkoxy groups represented by  $R_9$  and  $R_{10}$  in formula (3) have 1 to 20 carbon atoms, with C<sub>1-12</sub> alkoxy groups being more preferred. Specific examples of such alkoxy groups include methoxy, ethoxy, isopropoxy, t-butyloxy, and 2-ethylhexyloxy. Alkenyloxy groups that have 2 to 20 carbon atoms are preferred, and C<sub>2</sub>-C<sub>12</sub>-alkenyloxy groups are more preferred. Examples of the naphthalene groups which are represented by Ar in formula (1) and which may optionally have substituents include those which have one or two substituents such as alkoxy, alkenyloxy, hydroxy, acyl, alkyl, alkenyl, or substituted with



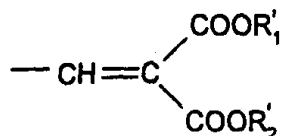
wherein  $R'_1$  and  $R'_2$  have the same meaning above for (xi). Specific examples of such alkenyloxy groups include allyloxy and 3-butenyloxy.

With reference to (3) it is preferred that the  $R_9$  acyl groups have 2 to 20 carbon atoms.

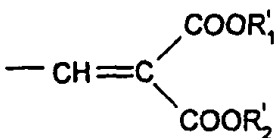
Among them, C<sub>2</sub>-C<sub>12</sub> acyl groups are more preferred. Specific examples of such acyl groups include acetyl, propionyl, and benzoyl.

The alkyl groups and alkenyl groups of (3) for R<sub>9</sub> or R<sub>10</sub> are preferably C<sub>1</sub>-C<sub>20</sub> alkyl groups and C<sub>2</sub>-C<sub>20</sub> alkenyl groups; and particularly, C<sub>1</sub>-C<sub>12</sub> alkyl groups and C<sub>2</sub>-C<sub>12</sub> alkenyl groups are more preferred. Specific examples of such alkyl groups and alkenyl groups include the groups which were mentioned in relation to R<sup>1</sup> and R<sup>2</sup>.

With reference to (3) it is particularly preferred that R<sub>9</sub> be a hydrogen atom, an alkoxy group or a hydroxy group. Also, it is particularly preferred that R<sub>10</sub> be a hydrogen atom, an alkoxy group, or a group of the formula

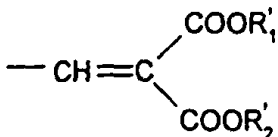


The group

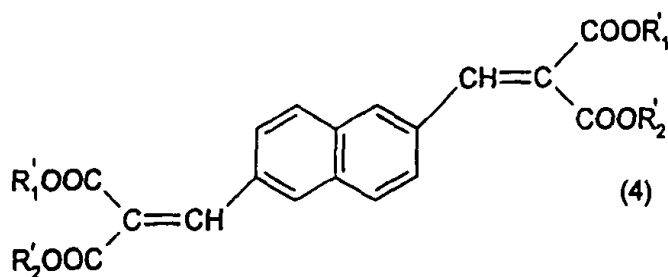


in formula (3) may be substituted at either the first or the second position of the naphthalene skeleton.

Also, among the compounds of formula (3) where R<sub>10</sub> is

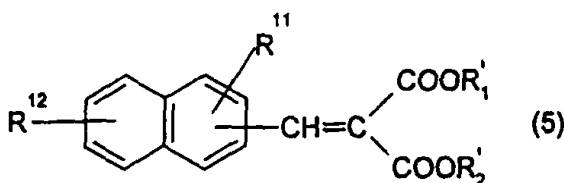


are thus represented by formula (4) and are particularly preferred examples in respect of (1) where Ar is a naphthyl group.



wherein  $R_1'$  and  $R_2'$  have the same meaning as defined above.

Among the compounds of formula (3) which are particularly preferred are:



wherein

$R_{11}$  represents a hydrogen atom, a  $C_1 - C_{20}$  alkoxy group or a hydroxy group;

$R_{12}$  represents a hydrogen atom, a  $C_1 - C_{20}$  alkoxy group; and

$R_1'$  and  $R_2'$  have the same meaning as defined above.

The method of producing the naphthalenemethylenemalonate esters is disclosed in U.S. Patent 5,508,025 incorporated herein by reference.

Naphthalenemethylenemalonate diesters are obtained by conventional condensation of (Knoevenagel condensation) an aromatic compound and a malonic diester with or without solvent and in the presence of a catalyst. Examples of suitable solvent for this reaction include benzene, toluene, xylene, tetrahydrofuran, 1,4-dioxane, and alcohols.

Examples of the catalyst include amines such as piperidine and pyridine; acids such as acetic acid, benzoic acid, zinc chloride, and titanium tetrachloride; carboxylic acid salts such as sodium acetate and ammonium acetate; and acid anhydrides such as acetic anhydride. They may be used in suitable combinations. This reaction is preferably carried out with heat while removing water generated from the reaction.

In the reaction when a formyl group is present in the naphthylaldehyde derivative, two mols of malonic diester are condensed to produce the compounds of formula (4).

The additive represented by the formula (1)-(5) can be added to wax in neat form or as is preferred, compounds such as (5) be carried by a suitable carrier. No restrictions are imposed on the carriers as long as they are inert with respect to the malonic diesters. The carriers may take solid, liquid, emulsion, foam, or gel forms. Examples of the carriers include water; alcohols; oils and fats such as hydrocarbons, fatty acid esters, higher alcohols, and silicone oils; fine powders such as starch and talc; and agents for aerosol jetting such as low-boiling point hydrocarbons and hydrocarbon halides.

The compound of formula (1) is added under stirring to molten wax during the formulation step at a temperature generally from 150°F to 350°F (65.5-176.6°C), more typically, at 180°F to 230°F (82.2-110°C), depending upon the melting point for (1), prior to processing into slabs, granules, chips, pellets, prills, bars and the like, or shaped articles may be formed directly from the molten formulated wax by conventional means for molding molten wax formulations. The means for molding candles includes the conventional candle forming methods, such as casting in a container as in a container candle, votive, or taper.

An effective final concentration of compound (1) in a solid, colored shaded article can range from 0.2% to 0.5%, preferably 0.1 to 0.3%. A concentrated premixed embodiment, consists essentially of a wax and (1) dissolved therein, where the amount of (1) is advantageously higher, for example, from 0.5% to 5% by weight, preferably 1.0% to 2.0% of (1) is contained on the weight of wax. The concentrate can be formulated as a natural color, or a colorant can be added. The following optional components may be included in a formulated wax compound according to the invention, such as fatty acids, fatty esters, ethylene-vinyl acetate polymer (VAM content <20%), rosin esters, white mineral oil, hydrogenated castor oil, antioxidant, e.g., BHT, or tocopherol, fragrance, e.g. vanillin, cinnamon, floral fragrance, and the like (See CTFA publications, at [www.ctfa.com](http://www.ctfa.com)); low m.w. PE (<10,000 m.w.), hydrogenated fatty oils, petrolatum, beeswax, among the many other conventional additives can be included in the formulated wax containing (1).

The solid, colored, wax-based article of the present invention can be shaped in a conventional manner which includes, but is not limited to, casting, injection molding, extrusion, cold forming, vacuum forming, blow molding, compression molding, and transfer

molding. The article may be formed from granular or in slab form, or molded or otherwise formed into any shape, size, or form.

Shaped candle articles include container candles, pillar candles, and the well known tapered candles, as well as colored multilayered tapers, and the like. An exemplary multilayered taper candle has an overdipped layer, with colorant and (1) contained in the overdipped layer. A preferred tapered candle comprises hydrogenated fat and compound (1). Container candles comprise the wax and colorant and (1), the shape is the container. Containers are provided from glass vessels; and metal, ceramic, or pottery containers, and the like. Another preferred candle comprises bees wax, colorant, and compound (1).

### EXAMPLES

Percentages are expressed on a weight basis.

#### EXAMPLE 1 Base Wax

Votive candle base wax commercially available as Dussek Campbell no. 5454 was made by blending 4 kg of 130°F (54.4°C) melt point Fully Refined Paraffin (CASRN 64742-43-4), 4kg of 150°F (65.6°C) melt point Fully Refined Paraffin (CASRN 64742-43-4), and 2 kg of 155°F (68.3°C) melt point  $\alpha$ -olefinic wax (CASRN 131459-42-2). The mixture was heated to 180°F (82.2°C) and agitated for 20 minutes.

#### EXAMPLE 1A Comparative Base Wax

- |        |   |
|--------|---|
| 93.99% | Paraffin wax (Shellwax 100)(m.p. 126°F (52.2°C))(C <sub>20</sub> -C <sub>50</sub> )   |
| 5%     | Bareco ® FR 5315 paraffin wax(m.p. 127°F (52.8°C))  |
| 1%     | C <sub>&gt;10</sub> $\alpha$ -olefin, free-radical polymerized wax, containing residual peroxide, and Iodine value=6 (Vybar® 103) and |
| 0.1%   | of propanedioic acid, [(4-methoxyphenyl)methylene]-, dimethyl ester (CASRN 7443-25-6) were melted together and mixed.                 |

This base mixture was colored using the dyes listed in Table 2, with the results of color aging shown after exposure to Xenon Worn Test method 16E using the AATCC gray scale rating 1-5.

**EXAMPLE 2**

1980 g of base wax 1 was mixed with 20 g of Rhodamine Red oil soluble dy and agitated for 20 minutes at 180°F (82.2°C).

**EXAMPLE 3**

1900 g of base wax 1 was mixed with 100 g of Oil Yellow R oil soluble dye and agitated for 20 minutes at 180°F (82.2°C).

**EXAMPLE 4**

1966 g of base wax 1 was mixed with 44 g of Oil Red oil soluble dye and agitated for 20 minutes at 180°F (82.2°C).

**EXAMPLE 5**

199.8 g of dyed wax from Example 2 was mixed with 0.2 g of propanedioic acid, [(4-methoxyphenyl)methylene]-, dimethyl ester (CASRN 7443-25-6) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

**EXAMPLE 6**

199.8 g of dyed wax from Example 3 was mixed with 0.2 g of propanedioic acid, [(4-methoxyphenyl)methylene]-, dimethyl ester (CASRN 7443-25-6) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

**EXAMPLE 7**

199.8 g of dyed wax from Example 4 was mixed with 0.2 g of propanedioic acid, [(4-methoxyphenyl)methylene]-, dimethyl ester (CASRN 7443-25-6) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.



**COMPARATIVE EXAMPLE 1**

199.8 g of dyed wax from Example 2 was mixed with 0.2 g of 2-(2-hydroxy-3,5-di-*t*-*tert*-amylphenyl)benzotriazole (CASRN 25973-55-1) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

**COMPARATIVE EXAMPLE 2**

199.8 g of dyed wax from Example 2 was mixed with 0.2 g of 2-hydroxy-4-*n*-octoxybenzophenone (CASRN 1843-05-6) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

**COMPARATIVE EXAMPLE 3**

199.8 g of dyed wax from Example 2 was mixed with 0.2 g of 2-(2-hydroxy-5-*t*-octylphenyl)-benzotriazole (CASRN 3147-75-9) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

**COMPARATIVE EXAMPLE 4**

199.8 g of dyed wax from Example 2 was mixed with 0.1 g of 2-hydroxy-4-*n*-octoxy benzophenone (CASRN 1843-05-6) and 0.1 g of 2-(2-hydroxy-5-*t*-octylphenyl)-benzotriazole (CASRN 3147-75-9) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

**COMPARATIVE EXAMPLE 5**

199.6 g of dyed wax from Example 2 was mixed with 0.4 g of 2-(2-hydroxy-3,5-di-*tert*-amylphenyl)benzotriazole (CASRN 25973-55-1) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votiv candl s were poured into steel molds, allowed to cool overnight, and remov d from the molds.

**COMPARATIVE EXAMPLE 6**

199.8 g of dyed wax from Example 3 was mixed with 0.2 g of 2-(2-hydroxy-3,5-di-tert-amyphenyl)benzotriazole (CASRN 25973-55-1) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

**COMPARATIVE EXAMPLE 7**

199.8 g of dyed wax from Example 3 was mixed with 0.2 g of 2-hydroxy-4-n-octoxybenzophenone (CASRN 1843-05-6) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

**COMPARATIVE EXAMPLE 8**

199.8 g of dyed wax from Example 3 was mixed with 0.2 g of 2-(2-hydroxy-5-t-octylphenyl)-benzotriazole (CASRN 3147-75-9) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

**COMPARATIVE EXAMPLE 9**

199.8 g of dyed wax from Example 3 was mixed with 0.1 g of 2-hydroxy-4-n-octoxybenzophenone (CASRN 1843-05-6) and 0.1 g of 2-(2-hydroxy-5-t-octylphenyl)-benzotriazole (CASRN 3147-75-9) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

**COMPARATIVE EXAMPLE 10**

199.8 g of dyed wax from Example 4 was mixed with 0.2 g of 2-(2-hydroxy-3,5-di-tert-amyphenyl)benzotriazole (CASRN 25973-55-1) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

**COMPARATIVE EXAMPLE 11**

199.8 g of dyed wax from Example 4 was mixed with 0.2 g of 2-hydroxy-4-n-octoxybenzophenone (CASRN 1843-05-6) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

**COMPARATIVE EXAMPLE 12**

199.8 g of dyed wax from Example 4 was mixed with 0.2 g of 2-(2-hydroxy-5-t-octylphenyl)-benzotriazole (CASRN 3147-75-9) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

**COMPARATIVE EXAMPLE 13**

199.8 g of dyed wax from Example 4 was mixed with 0.1 g of 2-hydroxy-4-n-octoxybenzophenone (CASRN 1843-05-6) and 0.1 g of 2-(2-hydroxy-5-t-octylphenyl)-benzotriazole (CASRN 3147-75-9) and agitated for 20 minutes at 180°F (82.2°C). Two 2" x 1.75" votive candles were poured into steel molds, allowed to cool overnight, and removed from the molds.

Tests were performed by placing a candle made with each color and additive into direct sunlight and under fluorescent light. Tests were also performed by placing a candle of each colored wax without an additive into direct sunlight and under fluorescent light. The samples were observed visually on weekly intervals to determine color fading. It was noted when initial color changes were first indicated and when any portion of the wax was completely devoid of color (i.e., bleached). The samples were studied over a twelve (12) week period. Table I lists the results of these tests.

**TABLE I Light Exposure Tests**

	Sunlight		Fluorescent	
	Color Change	Bleached	Color Change	Bleached
<b>Rhodamine Red Dye</b>				
Example 2-control	1 week	1 week	1 week	1 week
Example 5	5 weeks	8 weeks	None after 12 weeks	None after 12 weeks
Comp. Example 1	2 weeks	3 weeks	4 weeks	6 weeks
Comp. Example 2	2 weeks	3 weeks	4 weeks	6 weeks
Comp. Example 3	2 weeks	3 weeks	4 weeks	6 weeks
Comp. Example 4	2 weeks	3 weeks	4 weeks	6 weeks
Comp. Example 5	4 weeks	6 weeks	8 weeks	10 weeks
<b>Oil Yellow R Dye</b>				
Example 3-control	1 week	1 week	1 week	1 week
Example 6	3 weeks	3 weeks	10 weeks	10 weeks
Comp. Example 6	2 weeks	2 weeks	6 weeks	6 weeks
Comp. Example 7	2 weeks	2 weeks	6 weeks	6 weeks
Comp. Example 8	2 weeks	2 weeks	6 weeks	6 weeks
Comp. Example 9	2 weeks	2 weeks	6 weeks	6 weeks
<b>Oil Red Dye</b>				
Example 4-control	1 week	1 week	1 week	1 week
Example 7	6 weeks	9 weeks	None after 12 weeks	None after 12 weeks
Comp. Example 10	3 weeks	6 weeks	5 weeks	7 weeks
Comp. Example 11	4 weeks	6 weeks	6 weeks	8 weeks
Comp. Example 12	3 weeks	6 weeks	5 weeks	7 weeks
Comp. Example 13	4 weeks	6 weeks	6 weeks	7 weeks

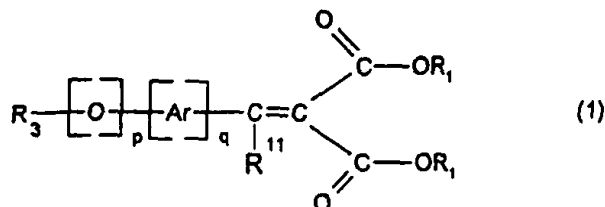
The series of examples in Table 2 illustrate the effect on UV exposure with and without the presence of propanedioic acid,[(4-methoxyphenyl)methylene]-, dimethyl ester (CASRN 7443-25-6). In most instances, the loss of color was more pronounced using the UVA in the presence of free-radical polymerized  $\alpha$ -olefin wax and paraffin wax.

**TABLE 2**

DYE	Level of UVA	Xenon exposure (hours) Grey Scale rating									
		(20)	(40)	(60)	(80)	(100)	(120)	(140)	(160)	(180)	(200)
S. Yellow 3G	None	5	5	5	4	3	3	3	3	3	3
"	0.1%	5	5	4	0	0	0	0	0	0	0
Host. Yellow 3G	None	3	2	1	0	0	0	0	0	0	0
"	0.1%	0	0	0	0	0	0	0	0	0	0
Solv. Yellow G	None	5	5	5	5	5	5	5	5	5	5
"	0.1%	5	4	4	4	3	3	2	1	1	1
Notro Fast Yell. B	None	5	5	5	5	4	4	4	4	3	2
"	0.1%	0	0	0	0	0	0	0	0	0	0
S. Orange 3G	None	5	5	5	5	5	3	3	3	3	3
"	0.1%	0	0	0	0	0	0	0	0	0	0
S. Orange G	None	5	5	5	5	5	5	4	4	3	3
"	0.1%	3	3	0	0	0	0	0	0	0	0
S. Red PFS	None	3	2	0	0	0	0	0	0	0	0
"	0.1%	1	0	0	0	0	0	0	0	0	0
Sando Blue 2B	None	5	5	5	5	5	5	5	5	5	5
"	0.1%	5	5	5	5	4	3	2	1	0	0
Sando Green G	None	5	5	5	5	5	5	5	5	4	4
"	0.1%	5	5	5	5	5	5	5	5	5	5
Sando Blue 2B	None	4	1	0	0	0	0	0	0	0	0
"	0.1%	5	5	5	5	4	3	2	1	0	0

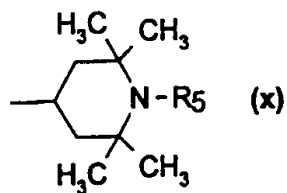
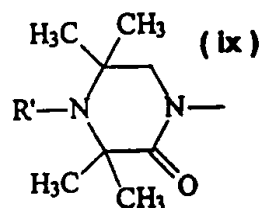
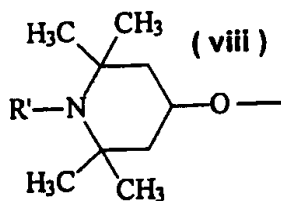
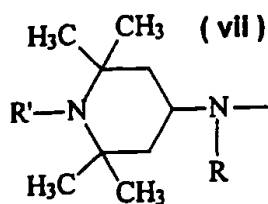
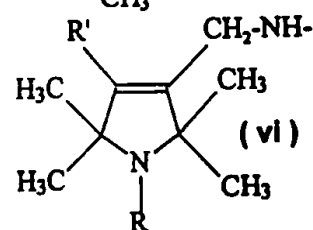
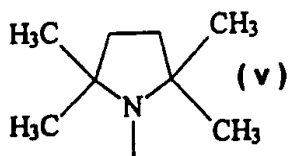
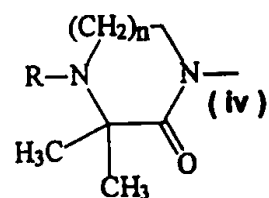
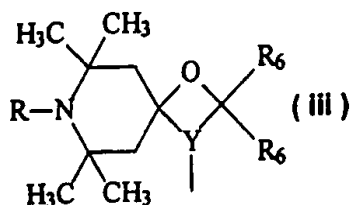
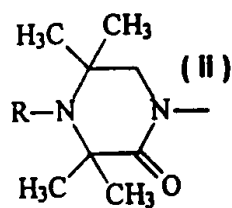
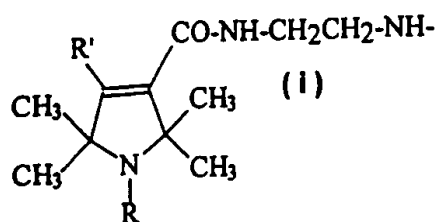
**WE CLAIM:**

1. A solid, colored, and shaped, wax-based article comprising: at least 75 weight % of natural, and/or synthetic wax, a colorant, and from 0.02% to 5% by weight based on the weight of said wax of a compound of the formula (1)

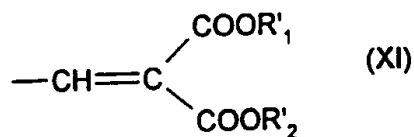


wherein

- $\text{R}_1$  is independently  $\text{C}_1$ -20 alkyl,  $\text{C}_1$ -20 alkoxyalkyl,  $\text{C}_1$ -20 hydroxyalkyl,  $\text{C}_1$ -20 alkenyl, substituted  $\text{C}_1$ -20 alkenyl groups,  $\text{C}_1$ -20-alkoxy- $\text{C}_1$ -20-alkyl groups,  $\text{C}_1$ -20-oxy-N- $\text{C}_1$ -20-alkyl groups, -N-cyclic- $\text{C}_1$ -10-alkyl groups, or cyclic-N- $\text{C}_1$ -10-alkyl groups;
- $\text{R}_{11}$  is hydrogen or  $\text{C}_1$ - $\text{C}_8$  alkyl;
- p and q are independently 0 or 1;
- Ar is a substituted aromatic single ring, an unsubstituted aromatic single ring, a substituted aromatic fused 2 or 3 ring group, an unsubstituted aromatic fused 2 or 3 ring group, a heteroaromatic single ring, or a heteroaromatic fused 2 or 3 ring group;
- $\text{R}_3$  is hydrogen,  $\text{C}_1$ - $\text{C}_{18}$  alkyl,  $\text{C}_5$ - $\text{C}_{12}$  cycloalkyl unsubstituted or mono-, di- or tri-substituted by  $\text{C}_1$ - $\text{C}_4$  alkyl, or  $\text{C}_7$ - $\text{C}_9$  phenylalkyl unsubstituted or mono-, di- or tri-substituted on the phenyl by  $\text{C}_1$ - $\text{C}_4$  alkyl, a hindered amino group selected from (i) to (x)



and a group (xi):



wherein

R, R', R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub> are independently either hydrogen, C<sub>1-12</sub>alkyl, C<sub>1-8</sub>alkoxy,  $r$ -COR<sub>3</sub>, where R<sub>3</sub> is hydrogen, C<sub>1-6</sub>alkyl, phenyl, -COO(C<sub>1-4</sub>alkyl) or NR<sub>15</sub>R<sub>16</sub>, where R<sub>15</sub> and R<sub>16</sub> are independently hydrogen, C<sub>1-12</sub>alkyl, C<sub>5-8</sub>cycloalkyl, phenyl or (C<sub>1-12</sub>alkyl)phenyl; or R<sub>15</sub> and R<sub>16</sub> together with the N-atom to which they are attached form a five- to seven-membered ring which may contain an additional N- or O-atom;

$n$  is 0 or 1 (structure iv);

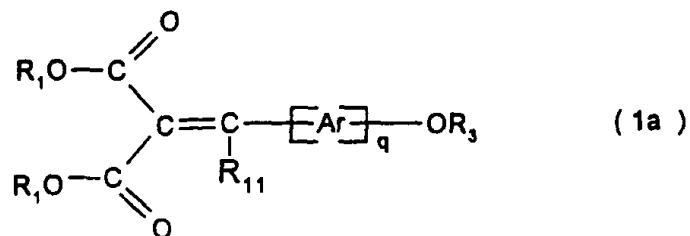
$Y$  is the group -NCO or -OCN, where CO forms part of the cyclic structure;

each R<sub>6</sub> of structure (iii) independently is selected from hydrogen, C<sub>1-12</sub>alkyl or phenyl provided only one R<sub>6</sub> can be phenyl, or both groups R<sub>6</sub> together form the group -(CH<sub>2</sub>) <sub>$m$</sub> -; where  $m$  is 2 to 11, -C(CH<sub>3</sub>)<sub>2</sub>-, -C(CH<sub>3</sub>)<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>- or -C(CH<sub>3</sub>)CH<sub>2</sub>CH<sub>2</sub>CH(CH<sub>3</sub>)-

2. The article as claimed in claim 1, wherein said compound (1) exhibits a UV absorptivity greater than or equal to 70 liters per gram-cm. at a wavelength of less than or equal to 400 nm.
3. An article as claimed in claim 1, wherein said compound formula (1) is present in an amount from 0.05 weight % to 1.0 weight % on weight of wax.
4. An article as claimed in claim 3, wherein said compound of the formula (1) is present in an amount from 0.1 weight % to 0.3 weight % on weight of wax.
5. An article as claimed in claim 1, wherein said wax comprises a Combination of paraffin wax and a monomeric or polymeric alpha olefin wax.
6. An article as claimed in claim 1, wherein said alpha olefin is an alkene C<sub>24</sub>-C<sub>54</sub>, branched or linear.



7. An article as claimed in claim 1, wherein said colorant is a dy .
8. A article as claimed in claim 1 wherein compound (1) is:



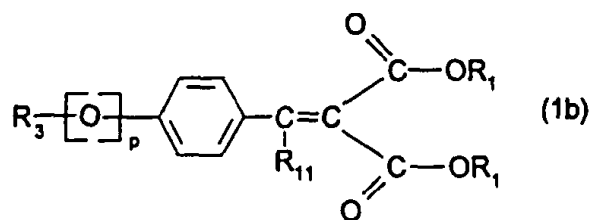
wherein

each R<sub>1</sub> is independently C<sub>1</sub>-20 alkyl, C<sub>1</sub>-20 alkoxyalkyl, C<sub>1</sub>-20 hydroxyalkyl, C<sub>1</sub>-20alkenyl, substituted C<sub>1</sub>-20 alkenyl groups, C<sub>1</sub>-20-alkoxy-C<sub>1</sub>-20-alkyl or selected from groups (l) to (x);

q is 1; and

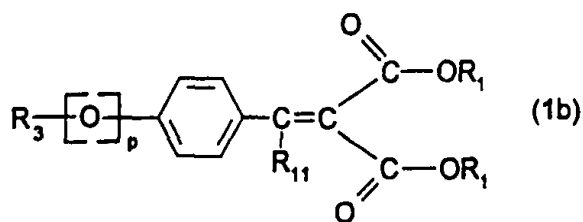
R<sub>3</sub>, R<sub>11</sub>, and Ar is as defined in Claim 1.

9. The article of claim 1 wherein compound (1) is



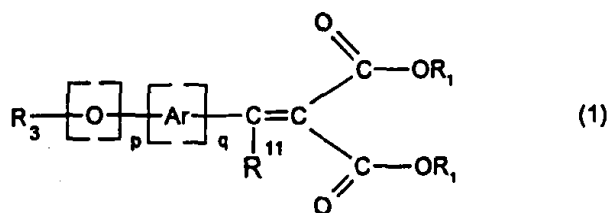
wherein p is 0 or 1, and R<sub>1</sub>, R<sub>3</sub>, R<sub>11</sub> are defined as in claim 1.

10. The article of claim 9 wherein compound (1b) is



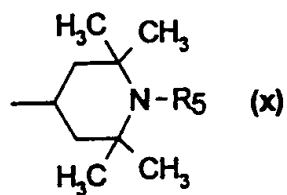
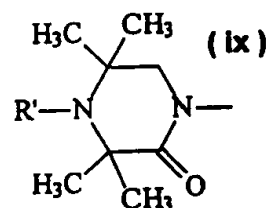
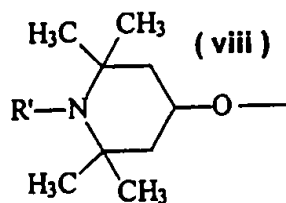
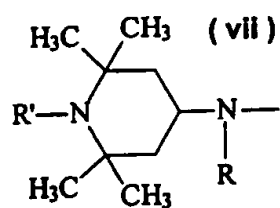
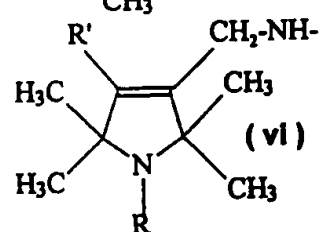
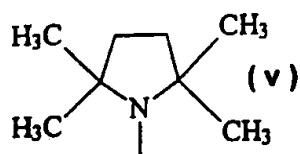
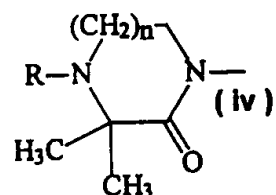
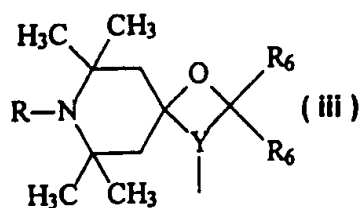
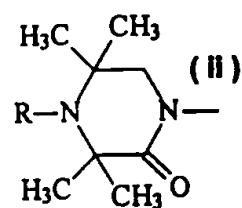
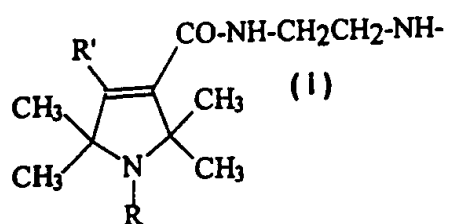
wherein  $R_1$  and  $R_4$  are linear or branched  $C_{1-6}$  alkyl groups.

11. A solid, colored, candle comprising: a natural and/or synthetic wax, a colorant, and a compound of the formula (1)

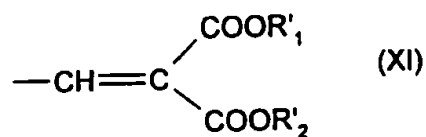


wherein

- $R_1$  is independently  $C_{1-20}$  alkyl,  $C_{1-20}$  alkoxyalkyl,  $C_{1-20}$  hydroxyalkyl,  $C_{1-20}$  alkenyl, substituted  $C_{1-20}$  alkenyl groups,  $C_{1-20}$ -alkoxy- $C_{1-20}$ -alkyl groups,  $C_{1-20}$ -oxy-N- $C_{1-20}$ -alkyl groups, -N-cyclic- $C_{1-10}$ -alkyl groups, and cyclic-N- $C_{1-10}$ -alkyl groups;
- $R_{11}$  is hydrogen or  $C_1-C_8$  alkyl;
- $p$  and  $q$  are independently 0 or 1;
- $Ar$  is a substituted or unsubstituted aromatic single ring or a substituted or unsubstituted aromatic fused 2 or 3 ring group or a heteroaromatic single ring or a heteroaromatic fused 2 or 3 ring group;
- $R_3$  is hydrogen,  $C_1-C_{18}$ alkyl,  $C_5-C_{12}$ cycloalkyl unsubstituted or mono-, di- or tri- substituted by  $C_1-C_4$ alkyl, or  $C_7-C_9$  phenylalkyl unsubstituted or mono-, di- or tri- substituted on the phenyl by  $C_1-C_4$ alkyl, or preferably a hindered amino group selected from (i) to (x),



or R<sub>3</sub> is a group (XI):



wherein

- R, R', and R<sub>3</sub> are independently either hydrogen, C<sub>1-12</sub>alkyl, C<sub>1-6</sub>alkoxy, or -COR<sub>3</sub>, where R<sub>3</sub> is hydrogen, C<sub>1-6</sub>alkyl, phenyl, -COO(C<sub>1-4</sub>alkyl) or NR<sub>15</sub>R<sub>16</sub>, where R<sub>15</sub> and R<sub>16</sub> are independently hydrogen, C<sub>1-12</sub>alkyl, C<sub>5-6</sub>cycloalkyl, phenyl or (C<sub>1-12</sub>alkyl)phenyl; or R<sub>15</sub> and R<sub>16</sub> together with the N-atom to which they are attached form a five- to seven-membered ring which may contain an additional N- or O-atom;
- n is 0 or 1;
- Y is the group -NCO or -OCN, where CO forms part of the cyclic structure;
- each R<sub>6</sub> of structure (iii) independently is selected from hydrogen, C<sub>1-12</sub>alkyl or phenyl provided only one R<sub>6</sub> can be phenyl, or both groups R<sub>6</sub> together form the group -(CH<sub>2</sub>)<sub>m</sub>-; where m is 2 to 11, -C(CH<sub>3</sub>)<sub>2</sub>-, -C(CH<sub>3</sub>)<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>- or -C(CH<sub>3</sub>)CH<sub>2</sub>CH<sub>2</sub>CH(CH<sub>3</sub>)-; and
- R'<sub>1</sub> and R'<sub>2</sub> in (xi) are selected independently from C<sub>1-20</sub> alkyl, C<sub>1-20</sub> alkoxyalkyl, C<sub>1-20</sub> hydroxyalkyl, C<sub>1-20</sub> alkenyl, substituted C<sub>1-20</sub> alkenyl groups, C<sub>1-20</sub>-alkoxy-C<sub>1-20</sub>-alkyl groups, C<sub>1-20</sub>-oxy-N-C<sub>1-20</sub>-alkyl groups, -N-cyclic-C<sub>1-10</sub>-alkyl groups, and cyclic-N-C<sub>1-10</sub>-alkyl groups.

12. A candle as claimed in claim 11, wherein said compound of the formula (1) is present in an amount from 0.02 weight % to 0.5 weight % on weight of wax in said candle.
13. A candle as claimed in claim 12, wherein said compound of the formula (1) is present in an amount from 0.1 weight % to 0.2 weight % on weight of wax in said candle.
14. A candle as claimed in claim 11, wherein said synthetic wax comprises an alpha olefin wax.
15. A candle as claimed in claim 11, wherein said alpha olefin is an alkene wax is C<sub>24</sub>-C<sub>34</sub>, branched or linear  $\alpha$ -olefin wax.

16. A candle as claimed in claim 11, wherein said colorant is an oil soluble dye.
17. A candle as claimed in claim 11 wherein said wax is bees wax.
18. A candle as claimed in claim 11 having a tapered elongated shape, and wherein said wax comprises hydrogenated fat.
19. The candle as claimed in claim 11 which contains an overdipped layer, in the shape of an elongated taper and wherein said colorant and (1) are contained in said overdipped layer.
20. A solid, colored, molded candle comprising: a polymerized alpha olefin wax, an oil soluble dye, and propanedioic acid, [(4-methoxyphenyl) methylene]-,dimethyl ester.
21. A candle as claimed in claim 21, wherein said propanedioic acid, [(4-methoxyphenyl) methylene]-,dimethyl ester is in an amount from 0.02 weight % to 0.5 weight % on weight of wax in said candle.
22. A candle as claimed in claim 21, wherein said propanedioic acid, [(4-methoxyphenyl) methylene]-,dimethyl ester is in an amount from 0.1 weight % to 0.3 weight % of wax in said candle.
23. A candle as claimed in claim 14, wherein said alpha olefin comprises an  $\alpha$ -olefin  $C_{24}$ - $C_{34}$ , branched or linear wax.

24. The Candle of claim 11 wherein said colorant is selected from the group consisting of:

C.I. Solvent Yellow 3, C.I. Solvent Green 3, C.I. Acid Black 429, C.I. Solvent Red 207, C.I. Phthalo Blue, C.I. Solvent Violet 13, C.I. Disperse Violet 17, C.I. Solvent Yellow 14, C.I. Solvent Blue 36, C.I. Solvent Yellow 33, C.I. Solvent Red 149, C.I. Solvent Yellow 56, C.I. Solvent Yellow 90, C.I. Solvent Red 26, C.I. Solvent Yellow 29, Solvent Red 24, C.I. Solvent Yellow 16, C.I. Solvent Yellow 72, C.I. Solvent Blue 104, C.I. Solvent Yellow 93, C.I. Solvent Yellow 98, C.I. Disperse Yellow 64, C.I. Solvent Yellow 24, C.I. Solvent Orange 60, C.I. Vat Red 41, C.I. Solvent Red 195, C.I. Solvent Red 111, C.I. solvent Red 179, C.I. Solvent Red 135, C.I. Solvent Red 63, C.I. Disperse Violet 26, C.I. Solvent violet 37, C.I. Solvent Green 28, C.I. Solvent Yellow 133, C.I. Solvent Yellow 164, C.I. Solvent Blue 122, C.I. Solvent Violet 49, C.I. Solvent Brown 122, C.I. Solvent Red 91, C.I. Solvent Red 164, and C.I. Solvent Blue 98.

## INTERNATIONAL SEARCH REPORT

Inter national Application No

PCT/IB 99/01650

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08K5/11 C08K5/134 C08L91/06 C08L91/08 C09G1/08  
C11C5/00 C09D191/06 C09D191/08

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08K C08L C09G C11C C09D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	FR 1 487 593 A (GEIGY) 18 October 1967 (1967-10-18) the whole document ----	
A	US 4 301 209 A (LORENZ DONALD H ET AL) 17 November 1981 (1981-11-17) cited in the application column 5, line 21-46 ----	
A	US 5 624 663 A (DEFLANDRE ANDRE ET AL) 29 April 1997 (1997-04-29) column 1, line 65 -column 3, line 35 -----	

☐ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

## \* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"Z" document member of the same patent family

Date of the actual completion of the international search

18 November 1999

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# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

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